



Comparison of particulate matter emission and soluble matter collected from combustion cigarettes and heated tobacco products using a setup designed to simulate puffing regimes

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ABSTRACT

An experimental setup was designed and optimized to collect particulate matter suspended in cigarettes smoke or in the aerosol generated during the use of heated tobacco products (HTPs). The setup is mainly based on a gas washing flask containing water where particulate matter and soluble compounds can be trapped, resembling interaction in the body. This system allows to perform puffing experiments in conditions similar to those of the Health Canada Intense puffing regime. In the study, cigarettes and Heets (in IQOS heater) from Philip Morris Products were used. Complementary characterization of this particulate matter has been extracted from experiments on a gas washing flask containing isopropanol. Total organic carbon analysis, laser diffraction, UV-vis spectrophotometry and transmission electron microscopy measurements have shown that cigarette smoke contains a large number and content of compounds soluble in water and isopropanol, together with solid particles generated during combustion. In contrast, negligible presence of non-combustion related particles was detected in Heets aerosol samples. These facts, together with the higher amount of water soluble compounds and higher number of polyaromatic hydrocarbons detected in the cigarettes smoke, allow to conclude that HTPs are less harmful than combusted cigarettes.

1. Introduction

Cigarette smoke has shown to be the cause of health problems like cardiovascular and chronic obstructive pulmonary diseases and lung cancer [1–3], and this has led to health authorities like the World Health Organization (WHO) [4,5] to promote measures to smoking cessation. However, the impact of the adopted measures is quite limited [6,7]. With the purpose of reducing the inhalation of Harmful and Potentially Harmful Chemicals (HPHCs) [8–10], alternatives to regular cigarettes, like different tobacco and nicotine-containing products, such as electronic cigarettes, snus, or heated tobacco products (HTPs), have been developed. This approach intends to be an alternative for adult smokers who would otherwise continue smoking regular cigarettes.

HTPs have become widely spread and unlike cigarettes, in which tobacco burns at 700–950 °C during puffs, the temperature during the use of HTPs is lower than 350 °C [9,11]. In conventional cigarettes, the high temperatures achieved in the smoking process lead to the formation of thousands of chemical products by different physico-chemical routes

(e.g. pyrolysis, pyrosynthesis, distillation, sublimation and condensation processes) [12,13]. In contrast, the HTPs use involves the thermal decomposition of organic compounds present in the tobacco and new species are formed by pyrogenesis and pyrosynthesis, without the generation of combustion compounds and soot particles [12,14]. Delivery of nicotine and tobacco flavours by HTPs could help consumers to replace smoking cigarettes or to make a combined use of both systems [12,15,16].

IQOS from Philip Morris International (PMI) is a product in the HTP market available in more than 53 countries (data for the first half of 2020 [17]). The system is based on tobacco sticks (called Heets), which consist of cast-leaf tobacco material containing glycerine, that is electrically heated by a temperature-controlled heating blade in the IQOS device [8].

PMI has demonstrated that combustion does not occur in Heets use [18], and that the amount of generated HPHCs is significantly lower than in cigarettes smoking [8,12,19–24]. Studies of several research groups have also proved that the amounts of harmful compounds like

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carbonyl compounds, PAHs, and nitrosamines are substantially lower in IQOS aerosols than in cigarettes smoke [9,12,16]. Moreover, cytotoxicity and genotoxicity of the IQOS aerosol have proved to be much lower than for cigarette smoke [12,25].

The cigarettes smoke consists of a mixture of vapours and gases in which soot particles are suspended [26]. This smoke is the result of high temperature tobacco pyrolysis and combustion, being soot particles the consequence of cracking or carbonization processes [27–29]. In contrast, at the relative low temperature of the IQOS use, only distillation and thermal degradation processes occur [30]. Pratte et al. found that, in contrast to what happens with regular cigarettes, combustion-related particles were not present in the mainstream aerosol generated by IQOS system [30,31].

However, the conclusions of previous works, in particular those related to tobacco companies, must be strongly supported by independent studies in order to definitively assess the significantly lower harmful character of heated tobacco products. Moreover, it is interesting to analyse what happens when the cigarettes smoke or the Heets aerosol interact with a water-rich medium like the one of biological (human) tissues. With this aim, the present study has focused on analysing the species retained or formed when the stream generated in cigarettes smoking or Heets use is driven to flow through a water reaction bed.

For this purpose, an experimental set-up that allows to simulate puffs has been designed and optimized. The collected samples were analysed using state-of-the-art analytical techniques: total organic carbon content (TOC), laser diffraction (LD), UV-vis spectroscopy, gas chromatography-mass spectrometry (GC-MS) and transmission electron microscopy (TEM, both bright field (BF) and high-angle annular dark-field (HAADF)) coupled to EDX analysis (TEM-EDX). The results of this study allow to compare relevant properties of the cigarettes smoking and Heets use mainstreams and to assess the relative harmful potential of both of them, in particular regarding the interaction with water-rich substances.

2. Experimental

2.1. Experimental setup design and optimization

The design of the experimental setup built in this work was inspired by the one reported by Horváth et al. [32,33] (Figure S1 in Supplementary Information), and it was modified to fit the current research interest. In general, the developed setup includes a pump to make the mainstream smoke or aerosol flow through a series of washing flasks containing water. Different setup configurations were tested, including one, two, or even three gas washing flasks, containing 250 mL water each one. The flasks were connected to each other in a row by rubber tubes, and an empty flask was placed at the end and connected to a

vacuum pump. Eight iterations were necessary to optimize the setup in order to simulate the HCI (Health Canada Intense) puffing regime (inhalation of 55 mL in 2 s (known as a puff), every 30 s; in realistic puffing experiments the flowrate ranges between 500 and 1500 mL/min [12]) and, finally, the best performance was achieved using only one washing flask. Note that to increase the contact time of the smoke/aerosol with the water trap, the glass tube which drives the smoke/aerosol into the water volume was capped with a holey rubber piece (0.6 mm holes were poked using a fine needle) to generate small bubbles. A drawing and some photographs of this setup can be seen in Figures 1A and 1B, respectively.

A mouthpiece (with a tube) was fitted to hold a cigarette or a Heet (see Figure 1A) and a two-way glass stopcock was included to enable the intermittent puffing. This glass device allows to select one of the two paths depicted on Figure 1A (the stopcock position that enables path 1 connects the extraction system with the mouthpiece and the position that enables path 2 isolates the tobacco product from the extraction system, which is then connected to an open way). Consecutive selection of paths 1 and 2 in suitable time intervals leads to puffing simulation.

The puffing flow rate was adjusted to a value consistent with realistic puffing experiments by fixing the appropriate vacuum conditions (the flow rate was determined by measuring the volume of water displaced in a graduated cylinder in a fixed time, Figure S2 in Supplementary Information). With a reduced pressure of approximately 50 mbar the smoke/aerosol flow rate was 960 mL/min. Finally, the experimental conditions were adapted to obtain puffs in accordance to the HCI regime (55 mL puff volume, 2 s puff duration, 30 s puff period).

2.2. Tobacco products and IQOS device

The following tobacco products, purchased in a local tobacco shop in Alicante, were used in this study: regular tobacco cigarettes (Marlboro Red, Philip Morris USA Inc., Richmond, VA) and Heets (amber box from Philip Morris Products S.A., Neuchâtel, Switzerland). The IQOS device (2.2 model) was supplied by Philip Morris Products S.A. (Neuchâtel, Switzerland). To use the Heet tobacco stick, it must be inserted in the IQOS holder, where it is heated by an electrically controlled heating blade [8].

2.3. Sample collection and characterization of collected samples

Puffing tests were carried out using 15 cigarettes or Heets in the following conditions: 55 mL puff volume, 2 s puff duration, 30 s puff period. The consumption time per cigarette was close to 7 minutes and the number of performed puffs was 13, while in the case of Heets the consumption time was about 6 minutes and the number of performed puffs was 11.

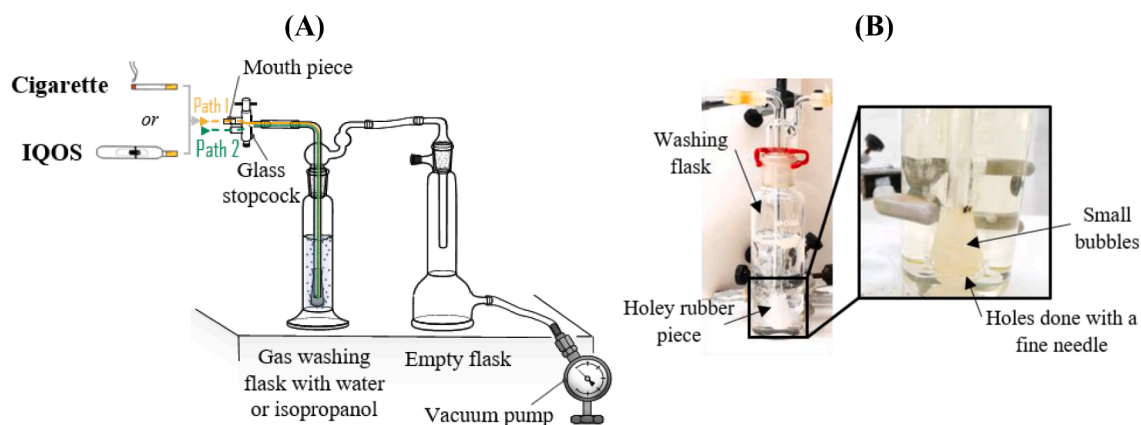


Figure 1. (A) Drawing of the complete experimental setup to perform puffing tests that simulate smoking or Heets use. (B) Photograph of the washing flask and detail of the holey rubber piece.

In some experiments isopropanol was used as trapping agent, instead of water, to trap as much as possible the smoke/aerosol suspended substances so as to have more information to compare the two studied systems. For analysis, 100 mL suspension/solution were collected from the gas washing flask.

The TOC of the filtered samples was determined by a Shimadzu TOC-V CSN equipment. Filtered solutions were also analysed by UV-Vis spectrophotometry using a UV-vis V-670 JASCO equipment. LD measurements were conducted using a Malvern Mastersizer 2000LF analyser (size working range 0.02–2000 μm in liquid media). GC-MS analysis was carried out in a low-resolution mass spectrometer with quadrupole analyser (Agilent 5975C) coupled to a gas chromatograph for capillary columns with split/splitless injector (Agilent 7890A). For TEM analysis, a drop of the collected sample was deposited on a grid holder and then the solvent was evaporated. TEM and TEM-EDX measurements were performed in a Thermo Scientific TALOS F200x model with energy dispersive X-ray spectroscopy signal detection. Particle size distribution was determined using the Jmol software and measuring the diameter of, at least, 100 particles.





3. Results and Discussion

Experiments with cigarettes led to yellowish solutions/dispersions in all cases, while in those for Heets the liquid remained colourless and clear (Table 1). The results of the analysis of these solutions/dispersions by several characterization techniques are presented next.

3.1. Total Organic Carbon (TOC) analysis

The obtained TOC results and the corresponding standard deviations calculated from quadruplicates for liquid samples obtained from tests using cigarettes or Heets were 553 ± 13 and 329 ± 28 mg/L, respectively. The obtained data indicate that TOC values in the collected water trap after cigarettes tests are about double than those after Heets tests. Taking into account that the consumption time is 20% higher for cigarettes than for Heets, TOC analysis allows to consider that, even using 20% more Heets, TOC content would be much higher in cigarettes samples. This difference would come from larger content of organic compounds, such as volatile organic compounds (VOCs), that can be partially dissolved in water. This is consistent with other authors claiming that cigarette smoke contains larger VOCs contents than the aerosol generated by the Heets [12,34].

Table 1
Photographs of the collected samples.

	15 cigarettes	15 Heets
Gas washing flask with water		
Gas washing flask with isopropanol		

3.2. Laser Diffraction (LD) analysis

As shown in Figure 2, the water sample from cigarettes smoking experiment contains particles of size ranging between 0.3 and 110 μm . The obtained particle size distribution in Figure 2 shows the main maximum at 15 μm , and two more relative maxima at about 0.7 and 100 μm . The particle sizes range for the samples collected in water trap for the cigarettes tests is larger than those for particles found in the mainstream smoke. Thus, some authors have found particles in the mainstream smoke with an average diameter of 75 nm, approximately [30, 35], although this mean size strongly depends on the number of puffs and the brand of cigarette used [35,36]. This size corresponds to ultra-fine particles found in combustion/pyrolysis processes [37]. The fact that particles in this work have larger mean size can be explained considering that they are suspended in water, so they can form condensates or aggregates. In the water samples collected in the 15 Heets tests no particles were detected by LD (six different samples were analyzed), meaning that either particles are not formed or their amount is below the detection limit of the equipment.

3.3. UV-vis spectrophotometry

After measuring UV-vis absorption spectra of water samples (W) and isopropanol samples (IP) collected from tests with 15 cigarettes or 15 Heets, the spectra of samples from cigarettes tests were too concentrated, leading to saturated signals, and they had to be diluted. Figure 3 shows the UV-vis absorption spectra for the different samples, indicating the dilution factor (DF) when used. In water samples from cigarettes, a DF of 7 (DF: total volume of the resulting solution divided by the volume of the concentrated solution) has allowed to notice absorption peaks centered at 210 and 260 nm (the second one as shoulder of the first one). A derivative analysis of this absorption curve (Figure S3 in supplementary information) reveals that it also seems to contain an overlapped shoulder at 290 nm. In the case of water samples from Heets tests, dilution was not necessary since the solution was much less concentrated, and two well defined peaks could be observed: a very intense incomplete one at 205 nm and a sharp one at 260 nm, with a shoulder at 290 nm. These results indicate that the flask water from both tobacco products experiments contains particular and relatively similar soluble compounds, present also in similar proportion in both samples, but not in similar concentrations. It is evident that the solutions from cigarettes tests are much more concentrated than the Heets ones (see Figure 3 inset, for comparison in concentration terms), meaning that the cigarettes smoke contains a much larger amount of water-soluble compounds that can be deposited on hydrated tissues. Isopropanol samples from the gas washing flask after cigarettes smoking experiments were also too concentrated for being characterized by this technique, and dilution was necessary. With 8.5 DF, the spectrum (see Figure 3) shows an intense peak located at 220 nm and a marked shoulder at 260 nm. The spectrum of the isopropanol solution from Heets experiments shows two well defined peaks of similar intensity at about 220 and 260 nm. In this case, the two isopropanol solutions also seem to contain similar compounds, although in different proportions, being the concentration much higher in the case of cigarettes samples.

These results highlight that the smoke from cigarettes and the aerosol from Heets contain more isopropanol-soluble compounds than water-soluble ones, and also that both isopropanol and water solutions from cigarettes tests are much more concentrated than those from Heets. Even using 20% more Heets to match the consumption times in both experiments, the UV-vis spectra intensity would hardly reach the obtained ones with 15 cigarettes. Thus, the concentration of dissolved compounds in both solvents is much higher in the cigarettes samples than in the Heets ones.

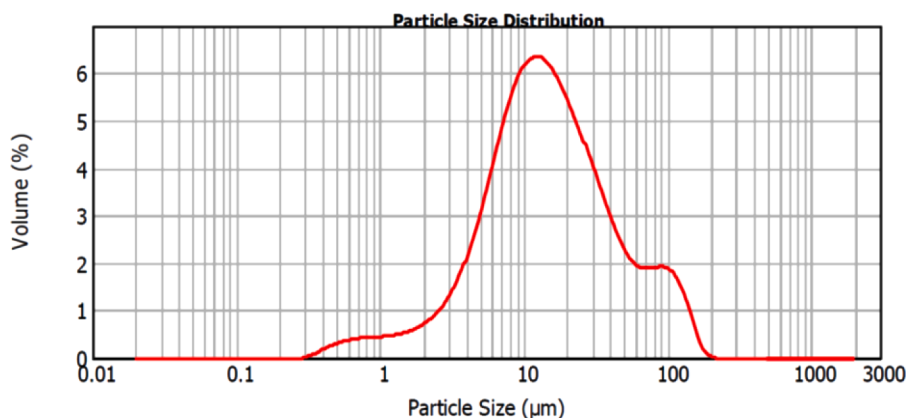


Figure 2. LD analysis of water from the flask collected after 15 cigarettes simulating puffs experiment.

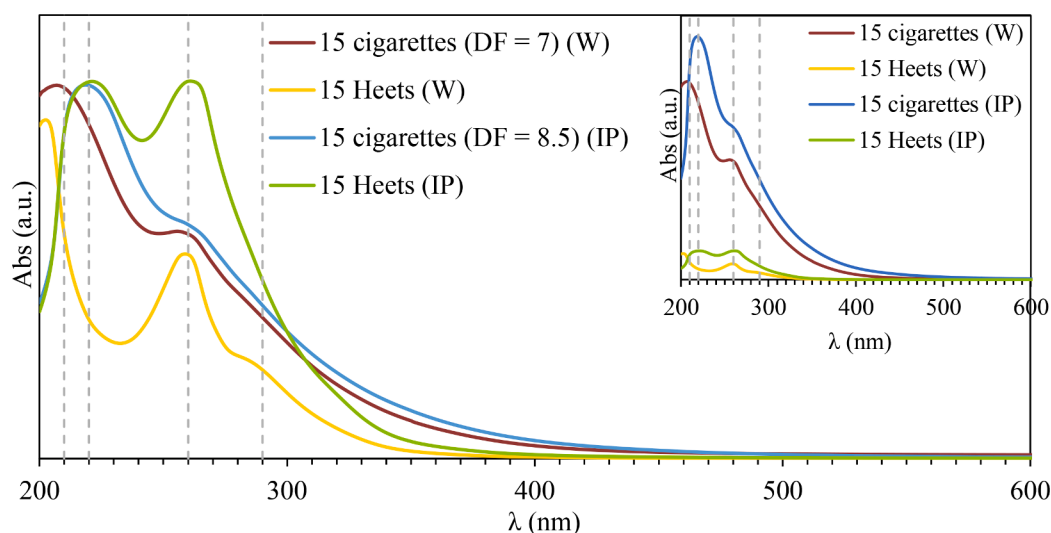


Figure 3. UV-vis absorption spectra of the filtered samples collected in water and isopropanol flask for different tests simulating puffs using 15 Heets or 15 cigarettes. Samples coming from cigarettes tests were diluted with different dilution factors (DF). Inset: UV-vis absorption spectra of the samples, including data for cigarette samples calculated multiplying the experimental data obtained for the diluted solutions by the DF, so as to achieve a quantitative comparison in terms of concentration.

3.4. Gas chromatography-mass spectrometry (GC-MS) analysis

Isopropanol samples collected from tests simulating puffs, involving 15 cigarettes or 15 Heets, were also analysed by GC-MS to detect the potential presence of polycyclic aromatic hydrocarbons (PAHs). In the cigarette samples, 40% out of the 29 identified compounds corresponded to PAHs (percentage obtained by integrating the chromatogram curves). In contrast, in Heets, approximately 2% of the 46 identified compounds corresponded to PAHs. Therefore, despite a greater variety of compounds was found in the isopropanol solutions for Heets experiments, only a very small portion were PAHs, part of HPHCs (harmful and potentially harmful constituents). On the contrary, for trapped cigarette smoke, fewer compounds were found, although a larger fraction of them was classified as HPHCs.

3.5. TEM and EDX analysis

Figure 4 shows representative bright field TEM images corresponding to samples collected after smoking 15 cigarettes using water (Figure 4 (a-f)) or isopropanol (Figure 4 (h-l)) as trapping solvents in the gas washing flask.

Images of Figure 4 (a-f), corresponding to samples collected in water, show well-defined homogeneous spherical structures, with a size distribution ranging between 30 and 180 nm and maximum at about 110 nm (Figure 4 (g)). The aspect of the samples from isopropanol solution (Figure 4 (h-l)) is very different and, in this case, a shapeless substance is

observed. The reason for such difference would be related with the composition of these structures and the media where they are collected. These solid structures have been either collected in water or in isopropanol, and some soluble organic fractions, especially in isopropanol, are condensed once the liquid media is evaporated during the grids preparation. Such a matter could be originated from compounds initially dissolved in isopropanol that precipitate upon solvent evaporation. These data highlight its organic nature and, according to the literature, it is composed by linear and aromatic hydrocarbons, aldehydes, esters and alcohols [38]. Note that this shapeless substance can be perfectly distinguished from an isopropanol insoluble part, which could be formed by combustion products with well-ordered and stable structures.

These results are in line with the higher solubility in isopropanol of cigarette smoke compounds, previously indicated, and they provide information about the shape and size of the solid matter that could be retained in water or aqueous media upon contact with cigarettes smoke.

These images (Figure 4 (a-l)) are very different from those corresponding to soot from fuel combustion processes [39], what can be explained considering that the composition of the cigarettes smoke and also the samples collection is very different.

Bright field TEM images of analogous samples collected from Heets experiments did not clearly show the presence of any solid matter, neither collected in water nor in isopropanol. In the case of water samples, and after a thorough search, very few areas were found to show structures that could be related to solid matter (Figure 5 (a-e)), but the amount can be considered negligible. TEM images of isopropanol

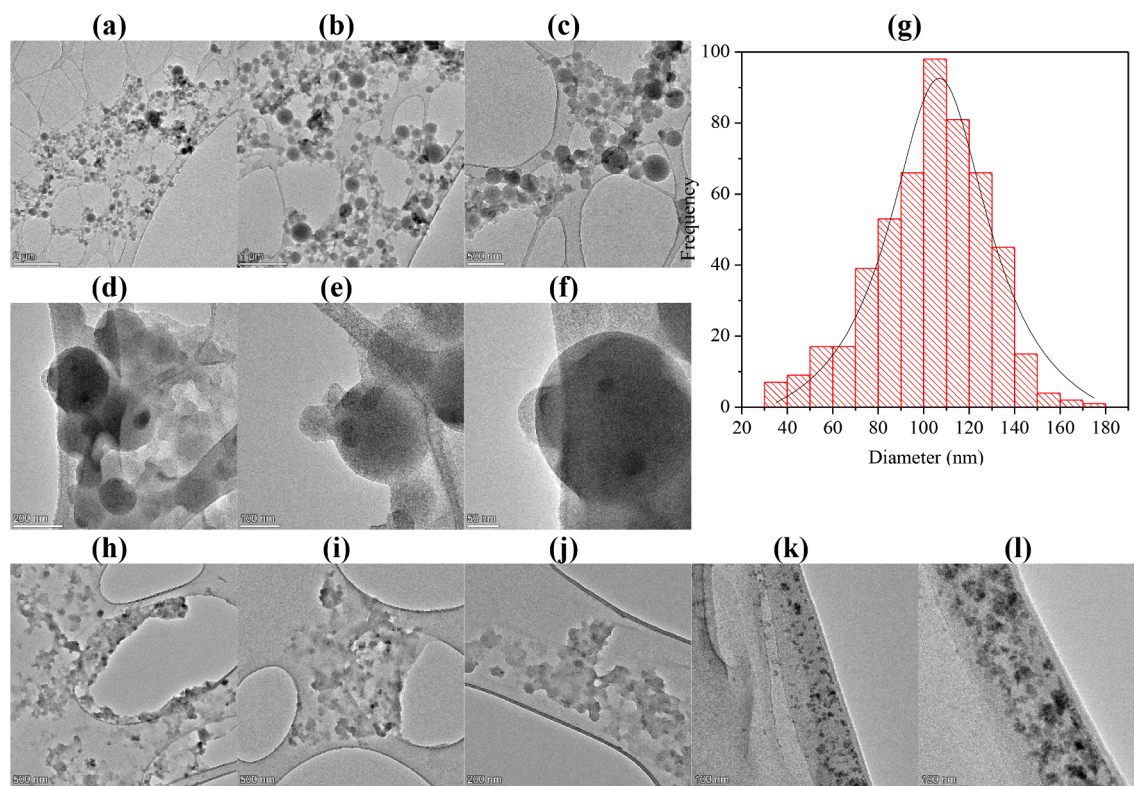


Figure 4. TEM images of particles found in water samples (a-f) and in isopropanol samples (h-l) collected in cigarettes smoking tests, and particles size distribution determined from cigarettes water samples (g).

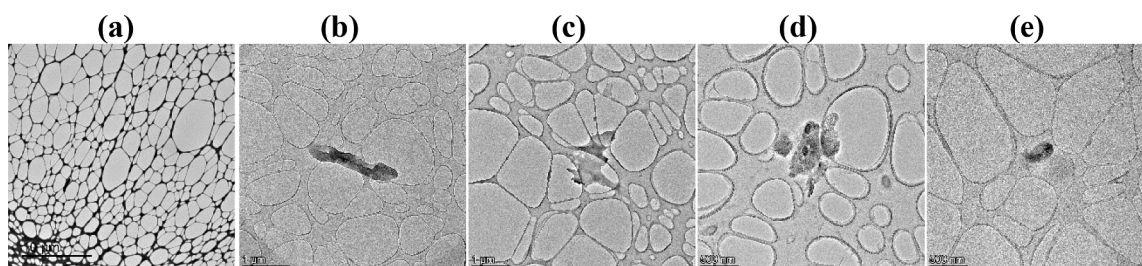


Figure 5. TEM images of water samples collected from 15 Heets simulating puffs experiments

samples did not show any solid matter at all, even after thorough search.

HAADF and EDX analysis was also carried out in the case of samples obtained from cigarettes smoking tests, both in water and in isopropanol (Figures 6a and 6b, respectively). HAADF images confirm, with high definition, the spherical structures detected in the water samples (Figure 6a). EDX analysis reveals that these particles mainly contain carbon (C), and oxygen (O), around 60 and 30 wt. %, respectively. Other elements like silicon (Si), potassium (K) and sulphur (S) are also present in appreciable amounts, being them all homogeneously distributed in the spheres.

The shapeless substance observed in the TEM analysis of isopropanol samples can be also distinguished in the HAADF images (Figure 6b). The chemical composition revealed by EDX shows that carbon (C) is the main element present (about 75 wt. %), oxygen (O) and silicon (Si) account for about 15 and 5 wt. %, respectively. Other elements detected in low amount are sulphur (S) and calcium (Ca).

A portion of the solid scarcely isolated in water from Heets experiments is shown in Figure 6c by HAADF TEM. This solid matter contains mainly carbon (C), around 90 wt.%, some oxygen (O), close to 10 wt.%, and very low amount of silicon (Si), sulphur (S) and calcium (Ca), being all these elements homogeneously distributed. TEM and TEM-EDX

analysis shows that the main differences between the water samples from cigarettes (Figure 6a) and Heets (Figure 6c) are: (i) particles population is much higher in the cigarettes samples than in Heets ones; (ii) cigarettes smoke leads to well defined spherical particles in water, while Heets aerosol generates an agglomerated structure with different shapes; (iii) the cigarettes sample is richer in O respect to the Heets one.

In summary, the differences in amount, composition and structure of the analyzed samples remark that cigarettes smoking produces harmful combustion particulate matter, while this does not happen in the case of HTPs use.

4. Conclusions

An experimental setup was designed, tested and optimized to simulate cigarettes smoking and HTPs use in a way that resembles the interaction of the produced mainstream with water (a media similar to that present in biological tissues) or with isopropanol (a good solvent for organic compounds, helpful to provide information about the nature and compositions of the compounds present). Collected samples were thereafter analysed by TOC, LD, UV-vis spectrophotometry, GC-MS, TEM and TEM-EDX.

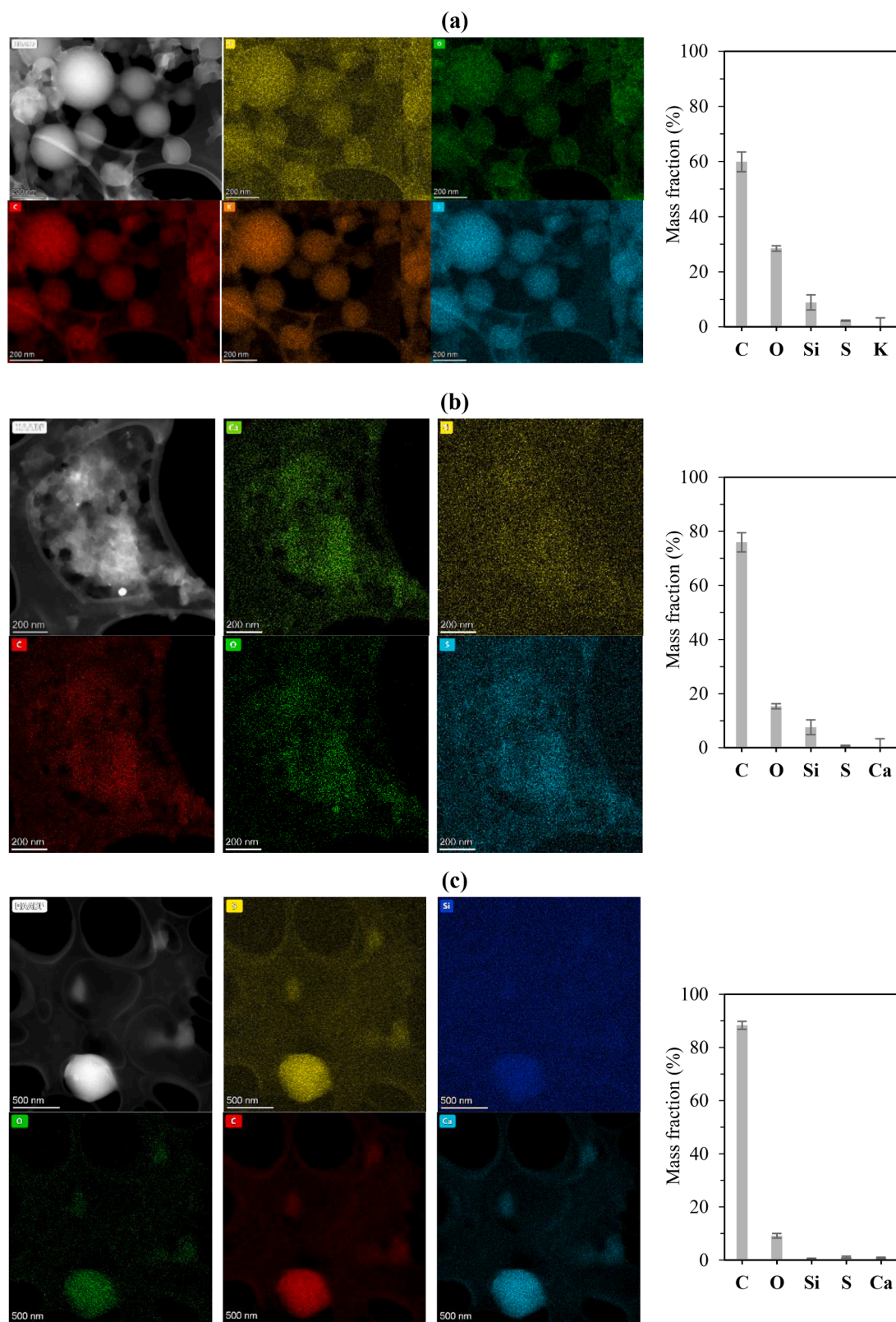


Figure 6. TEM-EDX characterization of a flask water sample (a) or a flask isopropanol sample (b), both from 15 cigarettes simulating puffs and of a flask water sample from 15 Heets simulating puffs test (c). Left: HAADF TEM image and TEM-EDX elemental mapping images. Right: Mass fraction values (in wt. %) identified by TEM-EDX of the elements in two portions of each sample.

The total organic carbon in water after cigarettes smoking is much higher (double amount) than the one from Heets aerosol. This is fully supported by the UV-vis absorption results, which also show that the type of compounds dissolved in water are of similar nature in both cases, cigarettes and Heets, although the content is much greater for cigarettes. Laser diffraction measurements have revealed the presence of particles ranging 0.3 - 110 µm in cigarettes samples, while no particles were detected in Heets samples. GC-MS analysis showed that in aerosol from Heets, HPHCs are a much lower fraction of the identified compounds than in the cigarettes smoke. TEM analysis has shown a high population of spherical particles in the cigarettes samples, while very few particles were detected in Heets samples. Isopropanol solutions/suspensions from cigarettes and Heets experiments also show important differences: the first are clearly more concentrated, as revealed by UV-Vis spectrophotometry, but the dissolved compounds seem to be different and present in different proportion. Besides, a shapeless substance, likely formed after dissolution and condensation, is observable in the cigarettes samples, but absent in the Heets ones.

All these findings highlight the important differences in the potential negative effects of cigarettes smoke and Heets aerosol when they are inhaled: the amount of water-soluble substances that could be deposited in the human tissues is significantly higher in cigarette-derived samples. In addition, the cigarettes smoke contains a much higher proportion of HPHCs in the identified compounds than the Heets aerosol. These results clearly show the important differences in the use of these two tobacco products and points to a lower harmful effect of Heets.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

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